

Application No. 10/565,542
Declaration Under 37 C.F.R. § 1.132
Docket No.: 283044US0PCT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:
Oliver HUTTENLOCH, et al.

GROUP: 1621

SERIAL NO: 10/565,542

EXAMINER: Y. K. R. CUTLIFF

FILED: January 23, 2006

FOR: A PROCESS FOR THE PREPARATION OF ACYLPHOSPHINES

DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Dr. Oliver Huttenloch who deposes and states that:

1. I am a graduate of University Dortmund, Germany and received my Ph. D. degree in the year 2001.
2. I have been employed by BASF SE, Ludwigshafen for 7 years as a Chemist in the field of Research/Production.
3. I have read and am familiar with the content of U.S. application serial number 10/565,542.
4. Specification page 10, Comparative Example 1, describes a process (NOT of the claimed inventive embodiments) for the attempted preparation of bis-(2,4,6-trimethylbenzoyl)-phenylphosphine:

Comparative Example 1 (NOT of the claimed inventive embodiments):

Under an inert atmosphere and with exclusion of moisture a dispersion of sodium (11.2 g, 0.50 mol) in toluene (200 ml) is produced by stirring with a standard blade stirrer with 1500 rpm at 105 °C resulting in sodium with a main particle size of 950 µm. The mixture is cooled to 30-35°C without stirring. Then the stirrer is started again with 300 rpm and chlorobenzene (2.93 g, 0.026 mol) is added dropwise without any visible effects. Then n-butanol (0.1 ml) is

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added and the mixture is heated to 40-50°C leading to a black coloured suspension. This suspension is heated to 100°C and P,P-dichlorophenylphosphine (20.4 g, 0.114 mol) is added dropwise at this temperature with exothermic behaviour. After the addition of $\frac{1}{4}$ of the whole amount of P,P-dichlorophenylphosphine the reaction stops to develop heat. At this point n-butanol (0.05 ml) is added and the exothermic reaction does not start again. The addition of n-butanol (0.05 ml) is repeated and the exothermic reaction starts again. All the rest of the addition of P,P-dichlorophenylphosphine stays exothermic. The resulting brown suspension is stirred at 95-105°C for 60 min. The mixture is cooled to 75°C and 2,4,6-trimethylbenzoylchloride (45.85 g, 0.251 mol) is added dropwise over a period of 30 min at this temperature. An exothermic reaction was observable only for the first 10% of the total amount of 2,4,6-trimethylbenzoylchloride. The suspension is stirred at 90°C for 90 min. The mixture was hydrolysed with water (200 ml) and the phases were separated. The product phase was analysed by ^{31}P -NMR. This showed the desired bis-(2,4,6-trimethylbenzoyl)-phenylphosphine only in traces (ca. 1%).

In contrast, specification pages 9-10 describe a process (of the claimed inventive embodiments) for preparation of bis-(2,4,6-trimethylbenzoyl)-phenyl phosphine. The phosphine was prepared as follows:

Example 1 (of the claimed inventive embodiments):

Under an inert atmosphere and with exclusion of moisture a dispersion of sodium (11.5 g; 0.50 mol) in toluene (100 ml) is produced by stirring with a high speed turbine stirrer with 11000 rpm at 105 °C resulting in sodium with a main particle size of less than 50 µm. The mixture is cooled to 30-35°C without stirring. Then the turbine stirrer is started again and chlorobenzene (2.8 g; 0.025 mol) is added dropwise and the contents of the flask warms to 45-50°C. The resulting black suspension is heated to 100°C and P,P-dichlorophenylphosphine (19.7 g, 0.11 mol) is added dropwise at this temperature with exothermic behaviour. After the addition of $\frac{1}{4}$ of the whole amount of P,P-dichlorophenylphosphine the reaction stops to

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develop heat. At this point n-butanol (0.05 ml) is added and the exothermic reaction starts again. All the rest of the addition of P,P-dichlorophenylphosphine stays exothermic. The resulting green suspension is stirred at 100-110°C for 30 min. The mixture is cooled to 75°C and 2,4,6-trimethylbenzoylchloride (43.8g; 0.24 mol) is added dropwise over a period of 30 min at this temperature. An exothermic reaction was observable. To the brown suspension was added toluene (200 ml) and the mixture was stirred for 60 min at 70-85°C. The mixture was hydrolysed with water (150 ml) and the phases were separated. The product phase was analysed by ^{31}P -NMR. This showed the desired bis-(2,4,6-trimethylbenzoyl)-phenylphosphine with 85-90% purity.

Additionally, specification pages 10-11, Example 2 (of the claimed inventive embodiments) describe a process for preparing bis-(2,4,6-trimethylbenzoyl)-phenylphosphine oxide:

Example 2: (of the claimed inventive embodiments):

Under an inert atmosphere and with exclusion of moisture a dispersion of sodium (11.5 g, 0.50 mol) in toluene (100 ml) is produced by stirring with a high speed turbine stirrer at 105 °C. With further stirring at 100°C, n-butanol (0.05 ml) is added. To the resulting grey suspension is added P,P-dichlorophenylphosphine (21.5 g, 0.12 mol) dropwise over a period of 20 min with constant development of exothermic heat. The mixture is then stirred for 30 min at 100-110°C and toluene (100 ml) was added. Then 2,4,6-trimethylbenzoylchloride (49.3 g, 0.27 mol) was added dropwise over a period of 30 min at 70-80°C. Only during the first 20% of the addition of 2,4,6-trimethylbenzoylchloride an exothermic reaction is observable. The reaction temperature is increased to 85°C and later to 110°C so to allow the exothermic reaction. The mixture is stirred at 110°C for 30 min. The temperature is lowered to 40°C and H_2O_2 (30%, 17.0 g, 0.15 mol) and water (150 ml) are added dropwise. The reaction is stirred at a temperature between 40 and 60°C for 2 h. The phases were separated. The product phase was analysed by ^{31}P -NMR. This showed the desired bis-(2,4,6-trimethylbenzoyl)-phenylphosphine oxide with 25% purity.

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Comments on Inventive Examples 1 and 2 and Non-Inventive Comparative Example 1:

In Comparative Example 1, the worked-up crude product, when analyzed by ^{31}P -NMR, demonstrated only traces of the desired product (ca 1%). Based on the combined teachings of Livingston, Leppard, and Stuebinger, the almost complete lack of desired compound in the crude reaction product constitutes the secondary consideration of recognition of a problem.

Additionally, as described in Example 1 and Example 2 (of the claimed inventive embodiments), respectively, after workup, an analysis of the crude isolated products revealed purities of from 85-90% and 25% respectively. These product purity results are superior to Comparative Example 1 (NOT of the claimed inventive embodiments, purity ca 1%). Further, based on the teaching of Livingston, Leppard, and Stuebinger, and as shown by Comparative Example 1 (NOT of the claimed inventive embodiments), these superior results are also unexpected results, thus demonstrating the secondary consideration of superior and unexpected results.

Without being bound by theory, the increased purities of crude worked-up reaction products of Examples 1 and 2, described above, correlate with, and are believed to at least in part result from, an increase in stirring speed. Example 1, for example, employed a turbine stirrer that stirred at 11,000 rpm. In contrast, and similar to Livingston where high speed stirring is conducted at 800-900 rpm (see column 4, Example 1 of Livingston), in Comparative Example 1, stirring speeds ranged from 300 to 1500 rpm. This correlation of significantly higher purities with higher speed stirring was surprising and unexpected based on the cited references. For example, one of ordinary skill in the art would necessarily understand, in reading, for example, Livingston, that 800-900 rpm would be expected to produce product in acceptable purities, yet as shown above, this is not the case. Thus, the correlation, and the superior purity results associated with higher stirring speed, are surprising and unexpected in light of the cited references.

5. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to

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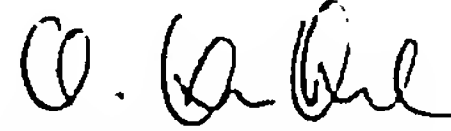
be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

6. Further deponent saith not.

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Signature

30.09 2009

Date